



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/EP87/00489 <b>(22) International Filing Date:</b> 21 August 1987 (21.08.87) <b>(31) Priority Application Number:</b> 8620341 <b>(32) Priority Date:</b> 21 August 1986 (21.08.86) <b>(33) Priority Country:</b> GB  <b>(71) Applicant (for all designated States except US):</b> HYDROGEN SYSTEMS N.V. [BE/BE]; Koolmijnlaan 201, B-3560 Beringen (BE). <b>(72) Inventor; and</b> <b>(75) Inventor/Applicant (for US only) :</b> VANDENBORRE, Jan-Baptist, Hugo [BE/BE]; Grootreesdijk 61, B-2460 Kasterlee (BE). <b>(74) Agent:</b> VAN REET, Staf; Populierenlaan 14, B-2460 Kasterlee (BE).		<b>(81) Designated States:</b> AT (European patent), BE (European patent), BR, CH (European patent), DE (European patent), FR (European patent), GB (European patent), IT (European patent), JP, NL (European patent), NO, SE (European patent), US.  <b>Published</b> <i>Without international search report and to be republished upon receipt of that report.</i>
<b>(54) Title:</b> BIPOLAR PLATE-SYSTEM FOR USE IN ELECTROCHEMICAL CELLS  <b>(57) Abstract</b> <p>A bipolar plate system for use in an electrochemical cell which comprises a non-conducting mechanical separation wall and extending therethrough electrically conducting elements connecting the working electrodes at both sides of the plate wherein said non-conducting plate is made of a thermosetting polymeric material.</p>		

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### BIPOLAR PLATE-SYSTEM FOR USE IN ELECTROCHEMICAL CELLS

This invention is concerned with electrodes, in particular a bipolar plate-  
5 electrode system for use in electrochemical cells.

Electrochemical cells presently in use for industrial purposes are mostly of  
the bipolar filter-press type. They comprise essentially a series of alterna-  
ting electrodes and ion-permeable membranes forming relatively thin  
anode and cathode compartments. One such compartment is comprised  
10 between an ion-permeable membrane at one side and an impermeable  
mechanical separation plate at the other side. Working electrodes, usually  
in the form of perforated metal plates, are kept in intimate contact with  
the ion-permeable membranes.

15 The working electrodes at both sides of the impermeable separation plate  
are electrically connected with each other and with the separation plate,  
which in the usual electrochemical cells is a metallic plate. Hence they are  
at the same potential. With respect to the preceeding and the subsequent  
electrode pair, they are at a higher, respectively lower potential and  
20 consequently one acts as an anode, the other as a cathode.

The overall construction of an electrochemical cell of this type is that of a  
filter-press and in general it is referred to as a filter-press-type bipolar  
cell. By means of an appropriate system of inlets and outlets, a constant  
25 flow of electrolyte fluid can be maintained in the anode and cathode  
compartments. The reaction products formed therein can be constantly

removed and transported to separate containers. In the case of water electrolysis, the electrolyte is an aqueous metal hydroxide solution, while hydrogen is produced at the cathode and oxygen at the anode.

- Intermixture of both gases must be absolutely prevented in order to  
5 preserve their chemical purity but also to avoid the formation of explosive oxygen-hydrogen mixtures.

In a usual construction, one unit of a filter-press type bipolar cell comprises the following elements:

10

- a. A semi-permeable separation wall, acting as an ion-permeable membrane which will allow the passage of ions and hence secure electrical conductivity while preventing the passage  
15 of the formed reaction products or reagents, in particular gases.

15

Materials currently used in semi-permeable separation walls comprise, for example, asbestos, inorganic materials such as oxides and hydroxides of various metals like Zirconium, Titanium, Antimony etc, organic wetting agents and organic polymers, nickel gauze, organic ion  
20 exchange polymers, etc.

20

- b. A working electrode which is kept in close contact with the ion-permeable separation wall. Usually the working electrode consists of a perforated or porous plate, made of an electrically conducting material  
25 which may optionally be covered with an electrocatalyst.

25

The electrode must be sufficiently inert under the often aggressive conditions wherein the cell is used (high concentration of acids or bases, high temperatures etc.) The predominantly used material for working electrodes in basic solution is nickel or nickel coated plated iron, while  
30 in acid solution electrodes of lead or lead oxide are generally preferred.

30

- c. A mechanical separation plate which separates an anodic and a cathodic compartment and which allows the working electrodes to be at the same potential. This plate is usually referred to as a bipolar plate.

In most of the filter-press electrochemical cells presently in use, the bipolar plates are made of metal. Electrical contact with the working electrodes is secured by inserting optionally flexible connecting elements between the bipolar plate and the working electrode. For optimal electrical contact, it is desirable to have the connecting elements, e.g. metal rods, fused or spotwelded on the bipolar plate and the working electrode. In a different set-up, the bipolar plate itself is pressed to take a tri-dimensional form with cams or points protuberating towards the working electrode at both sides of the bipolar plate.

In U.S. Patent Specification No. 3,849,279 there is described an electrolytic filter-press cell for the production of chlorine from aqueous alkali metal chloride solution wherein the metal anode and cathode of adjacent cells are in direct electrical connection with each other and said anode and cathode are maintained in spaced relationship by an electrically inert cell wall or barrier between them. According to the disclosure, the electrically inactive cell wall or barrier is made of a thermoplastic material, in particular a polyolefine. In order to prevent electrolyte and gas flow through the barrier, the metal rod connectors provided with a valve metal circumferential restraining flange, positioned substantially equidistant from each end of the said rod. The latter aspect renders the production of such cells both technically complicated and expensive.

In German Offenlegungsschrift No. 2600345, there are disclosed filter-press type electrolytic cells wherein the separation walls between two units consist of an electrically insulating material which is further specified as being e.g. a plastic or asbestos cement. In practice, plastics turn out to possess insufficient strength, particularly at elevated temperatures while asbestos cement is chemically not sufficiently stable.

d. Periferal structural and packing material containing openings which provide an in- and outlet system whereby, in the case of electrolytic

cells, fresh electrolyte can be pumped in constantly and a mixture of anodic respectively cathodic reaction products (gases) with electrolyte flows out into separate collectors.

- 5 Till now, none of the known constructions of bipolar plates are completely satisfactory. Major shortcomings have in general to do with their complex construction, the excessive use of expensive materials, insufficient mechanical strength or sealing capacity, and/or difficult assembly and disassembly of the cells.

10

By the present invention there is provided a particular construction of a bipolar plate for use in an electrochemical cell which is very simple in construction, inexpensive, efficient in operation and which has excellent mechanical strength and sealing capacity. Essentially the invention consists  
15 herein that, the mechanical separation wall is made of mechanically and chemically stable thermosetting polymer containing a set of electrically connecting elements extending to both sides of the plate and establishing adequate contact with the adjacent working electrodes.

- 20 In a preferred embodiment, the outer region of the bipolar plate is shaped such as to form at the same time a packing element, containing appropriate openings for the in- and outlet system.

As a thermosetting polymeric material to produce the mass of the bipolar  
25 plate, there can be used any type of thermosetting polymeric material which has sufficient mechanical strength and is sufficiently chemically inert under the conditions of operating the cell, and which in the unpolymerised form has appropriate flowability to adequately seal around the electrically connecting elements. Examples of such thermosetting  
30 materials are phenol-formaldehyde resins, aminoplasts like urea-formaldehyde and melanine-formaldehyde resins, tridimensional polyesters including alkyd resins and unsaturated polyesters, and epoxy resins.

- 35 Amongst the foregoing, epoxy resins are particularly preferred. For example, excellent results have been obtained with epoxy resins made

available by Emerson and Cumings under the tradename STYCAST 2651 MM using catalyst 9 or 11.

5 In principle the thermosetting materials can be incorporated into the bipolar plate under the molding process or they may be inserted afterwards in holes spared or cut out in the pre-molded plate. Preferably the connecting elements are incorporated into the plate during the molding process since this is the easiest and most effective way of securing a tight closure around the connecting elements.

10

It has indeed been found that, unlike with other construction elements such as thermoplastic materials, thermosetting materials exert a much more efficient and irreversible closure around the electrically connecting elements, thus securing the absence of any undesirable leaks without a  
15 need for special measures. Consequently, the material lends itself especially for the construction of cost-efficient electrolytic cells.

The connecting elements may take any form which allows a suitable electrical connection between the working electrodes while leaving  
20 sufficient space for the flow of electrolyte through the electrode compartments. In a simple and convenient embodiment, the connecting elements consist of cylindrical or other prismatically shaped rods extending through the separation plate. If desired this profile may be somewhat modified, e.g. to increase the surface area which is in contact  
25 with the working electrodes or to prevent sliding of the rods in the mass of the separation plate.

The connecting elements may be constituted of one single piece and, optionally, more than one or even all of the connecting elements of a  
30 particular separation plate may be unified in a particular tridimensional structure which lends itself to incorporation into the polymeric mass of the separation plate. Alternatively, a particular connecting element may be composed of more than one piece, joined together by conventional means, e.g. by screwing, clamping, rivetting etc.

The connecting elements may be substantially inflexible (e.g. in the case of rods) or they may have a more flexible structure in order to adapt themselves better to small structural variations which might otherwise give rise to a less than optimal contact between adjacent elements of an electrochemical cell. Such may be achieved, for example, by using bent connecting elements made of flexible metal, e.g. strings or angular elements or by making at least part of the connecting element of a relatively soft deformable metallic material.

10 In a preferred embodiment of the invention, the peripheral region of the separation plate is made thicker than the central area, making it essentially equiplanar with the surface of the working electrodes. With the right shape and size and compressibility of the separation plate it is possible to use minimal or even no packing material or frame to secure  
15 tight contact between the central and peripheral region of the bipolar electrode and the ion-permeable membrane. However, in order to improve contact between the peripheral region of the bipolar plate and the ion-permeable membrane it may be useful to hold the membrane in a somewhat thicker ring of a suitable material which combines adequate  
20 mechanical strength and rigidity with appropriate packing properties.

In a particularly preferred embodiment, the central and peripheral regions of bipolar plates and ion-permeable membranes fit onto each other in such manner that no additional packing material is required to produce a  
25 leakage-proof electrochemical cell. In principle the central and peripheral region of a bipolar plate can be made of different materials. For example, it may be advantageous to use a more rigid material for the central region and a more flexible material for the peripheral ring. Preferably, both are made of the same structural material.

30

In order to enable a constant flow of electrolyte into the individual electrolytical cell compartments and separate outlets for anolyte and catholyte, the peripheral parts of the bipolar plates and membranes must be foreseen with appropriately positioned openings which, taken together  
35 form an inlet and outlet system.



A schematic representation of a bipolar plate electrode system according to the invention will be found in Figures I (frontview) and II (section) wherein 1 is a bipolar plate, made of a thermosetting resin, 2 are electrically conducting studs (rods) extending through the bipolar plate and kept in contact with the perforated electrodes 3; the element 4 is the peripheral structural ring containing springs 5 and 6 for the in- and outlet system.

The bipolar plates according to the invention can be used in 10 electrochemical cells for use in various applications e.g. electrolytic cells and fuel cells or batteries. Examples of electrolytic cells are cells for the production of chlorine and particularly water electrolysis cells where hydrogen is formed in the cathodic and oxygen in the anodic compartments. The Figures attached to this description are meant to 15 illustrate and not to limit the scope of the invention.

CLAIMS

- 1 1. A bipolar plate for use in an electrochemical cell comprising a  
2 non-conducting separation plate and extending therethrough conducting  
3 elements making electrical contact with the working electrodes at both  
4 sides of the supporting plate, characterized in that the said  
5 non-conducting separation plate is made of a thermosetting polymeric  
6 material.  
7
- 8 2. A bipolar plate according to claim 1 wherein the said non-conducting  
9 separation plate is made of an epoxy resin.  
10
- 11 3. A bipolar plate according to any one of claims 1 and 2 wherein the said  
12 connecting elements are essentially prismatic metallic rods.  
13
- 14 4. A bipolar plate according to any one of claims 1 and 2 wherein the said  
15 connecting elements are flexible metallic elements.  
16
- 17 5. A bipolar plate according to any one of claims 1 to 4 for use in a the  
18 hydrolysis of water.  
19
- 20 6. A bipolar plate according to any one of claims 1 to 4 for use in the  
21 production of chlorine.

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Section A-B

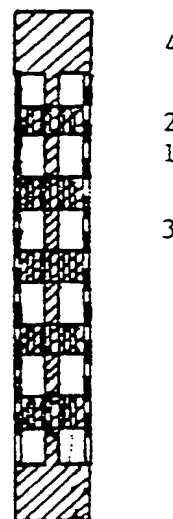


Figure I

Figure II

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<b>(54) Title:</b> BIPOLAR PLATE-SYSTEM FOR USE IN ELECTROCHEMICAL CELLS  <b>(57) Abstract</b>  A bipolar plate system for use in an electrochemical cell which comprises a non-conducting mechanical separation wall and extending therethrough electrically conducting elements connecting the working electrodes at both sides of the plate wherein said non-conducting plate is made of a thermosetting polymeric material.		

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# INTERNATIONAL SEARCH REPORT

International Application No.

PCT/EP 87/00489

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (If several classification symbols apply, indicate all) <sup>6</sup> According to International Patent Classification (IPC) or to both National Classification and IPC IPC <sup>4</sup> :                      C 25 B 9/04; H 01 M 8/02																							
<b>II. FIELDS SEARCHED</b> <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Minimum Documentation Searched <sup>7</sup></div> <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 25%; border-bottom: 1px solid black;">Classification System</td> <td style="border-bottom: 1px solid black;">Classification Symbols</td> </tr> <tr> <td style="height: 40px; vertical-align: top; border-right: 1px solid black;">IPC<sup>4</sup></td> <td style="vertical-align: top;">C 25 B 9</td> </tr> </table> <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched <sup>8</sup></div>			Classification System	Classification Symbols	IPC <sup>4</sup>	C 25 B 9																	
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IPC <sup>4</sup>	C 25 B 9																						
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT <sup>9</sup></b> <table style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 10%; border-bottom: 1px solid black;">Category <sup>9</sup></th> <th style="width: 70%; border-bottom: 1px solid black;">Citation of Document, <sup>11</sup> with Indication, where appropriate, of the relevant passages <sup>12</sup></th> <th style="width: 20%; border-bottom: 1px solid black;">Relevant to Claim No. <sup>13</sup></th> </tr> <tr> <td style="text-align: center; vertical-align: top;">X</td> <td style="vertical-align: top;">EP, A, 0002268 (ALLIED CHEMICAL CORP.) 13 June 1979 see page 7, lines 2-12; figures 3,4</td> <td style="text-align: center; vertical-align: top;">1,6</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td style="text-align: center; vertical-align: top;">--</td> <td style="text-align: center; vertical-align: top;">4</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td style="vertical-align: top;">DE, C, 563393 (I.G. FARBENINDUSTRIE) 6 February 1929 see figure 3</td> <td style="text-align: center; vertical-align: top;">4</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">P,X</td> <td style="vertical-align: top;">US, A, 4670123 (THE DOW CHEMICAL CO.) 2 June 1987 see column 3, lines 45-68; column 4, lines 1-13</td> <td style="text-align: center; vertical-align: top;">1-3,6</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">P,X</td> <td style="vertical-align: top;">EP, A, 0229473 (ICI) 22 July 1987 see column 8, lines 39-43; column 8, lines 8-9; columns 16-18, claims; figures 1,2</td> <td style="text-align: center; vertical-align: top;">1,2,4,6</td> </tr> <tr> <td colspan="3" style="text-align: center; padding-top: 20px;">-----</td> </tr> </table>			Category <sup>9</sup>	Citation of Document, <sup>11</sup> with Indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>	X	EP, A, 0002268 (ALLIED CHEMICAL CORP.) 13 June 1979 see page 7, lines 2-12; figures 3,4	1,6	Y	--	4	Y	DE, C, 563393 (I.G. FARBENINDUSTRIE) 6 February 1929 see figure 3	4	P,X	US, A, 4670123 (THE DOW CHEMICAL CO.) 2 June 1987 see column 3, lines 45-68; column 4, lines 1-13	1-3,6	P,X	EP, A, 0229473 (ICI) 22 July 1987 see column 8, lines 39-43; column 8, lines 8-9; columns 16-18, claims; figures 1,2	1,2,4,6	-----		
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EP 8700489  
SA 18375

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		CA-A- 1112208	10-11-81
		JP-A- 60255989	17-12-85
DE-C- 563393		US-A- 1907812	
US-A- 4670123	02-06-87	None	
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		JP-A- 62156284	11-07-87
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